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Published in:
Physical Review E. Statistical, Nonlinear, and Soft Matter Physics

Link to article, DOI:
10.1103/PhysRevE.62.2992

Publication date:
2000

Document Version
Publisher's PDF, also known as Version of record

COMMENTS

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Comment on ''Flow-distributed oscillations: Stationary chemical waves in a reacting flow''

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(Received 19 October 1999)

In a recent paper by Kærn and Menzinger [Phys. Rev. E **60**, R3471 (1999)] a successful verification of the stationary space-periodic structures predicted by Andresén *et al.* [Phys. Rev. E **60**, 297 (1999)] was reported. Kærn and Menzinger suggest a mechanism for the formation of such structures that yields a linear relationship between the selected wavelength and the flow rate. We find this mechanism too simple and produce numerical simulations that support the original interpretation of these structures.

PACS number(s): 82.40.Bj, 47.20.Ky

In the presence of an open flow, the behavior of spatially extended unstable systems crucially depends on whether the instability is convective or absolute. Perturbations applied at the boundary of a convectively unstable state can penetrate the system, which then acts as a nonlinear filter and a spatial amplifier [1]. In this framework, it has been predicted theoretically [2] that stationary space-periodic structures may be formed in such open flow systems when the flow velocity exceeds a critical value ν_T, and a permanent displacement away from a Hopf unstable homogeneous steady state is introduced at the inlet. This mechanism is to be contrasted with other pattern-forming instabilities in reaction-diffusion systems that require either a differential diffusion process (Turing instability) [3] or a differential flow [4].

In [2], the mechanism leading to the formation of stationary space-periodic structures was compared to the Cherenkov effect observed when a charged particle moves in a transparent medium. In our one-dimensional case, the perturbation at the inlet acts as a particle moving at velocity $-\nu$ resulting in waves (radiation) to be ''emitted'' at frequency ω. The stationary case arises as the phase velocity of the ''emitted'' waves and the flow velocity ν are equal:

$$\frac{\omega(k)}{k}=\nu. \tag{1}$$

For the model studied in [2], the selected wave number was correctly predicted by using the dispersion relation given by the weakly nonlinear description of the system.

Recently, Kærn and Menzinger [5] have reported a successful experimental verification of this phenomenon in a tubular reactor. Furthermore, they have proposed a mechanism whereby the stationary structures are the result of oscillating subelements, with oscillation period T, being transported downstream by the flow. The constant displacement at the inlet fixes the phase and should therefore yield stationary structures with a wavelength that obeys the following *universal* linear law: $\lambda=\nu T$. We think that this explanation, however attractive, is too simple as it does not take into account either the convective nature of the state or the criterion for amplifying waves. Both of these criteria must be fulfilled in order to observe stationary structures as only transient or evanescent waves will occur otherwise. Furthermore, Eq. (1)

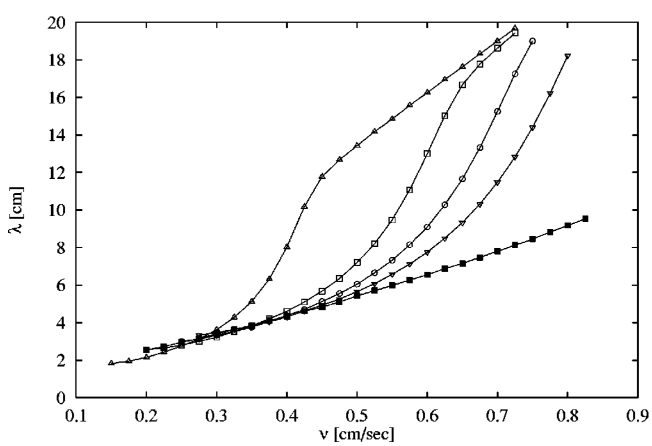

FIG. 1. Measured wavelengths from numerical simulations of the RZ model under different conditions for the diffusion coefficient. With the same parameters as in the experiments by Kærn and Menzinger [5] and $D=0.3\nu$, we observe approximate linear behavior (■). For fixed diffusion coefficient, $D=0.03$ (△), $D=0.06$ (□), $D=0.08$ (○), $D=0.1$ (▽), the wavelength does not exhibit a linear dependence on the flow velocity. Note that the curves start at the flow-velocity threshold ν_T for stationary space-periodic structures to occur.

generally yields a nonlinear relationship between the wavelength and the flow velocity that depends crucially on the nature of the bifurcation.

To illustrate our point, we have solved numerically the three-variable Rovinsky-Zhabotinsky (RZ) model [6] that describes well the kinetics of the ferroin-catalyzed Belousov-Zhabotinsky reaction used in the experiments (see Fig. 1). Our simulations show that for constant diffusion coefficient the wavelength grows with the flow velocity and only seems to relax to a linear dependence for high flow velocities. This behavior proceeds from the weakly subcritical Hopf bifurcation exhibited by the RZ model. On the other hand, if the diffusion coefficient depends linearly on the flow velocity $D = c\ \nu$ (turbulent diffusion), where c is some constant, then the selected wavelength may be perceived to depend linearly on the flow velocity for all velocities.

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